The Air Quality in Danish Urban Areas

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The Danish air pollution abatement is based by and large on emission control. Since the ratification of the international sulfur protocol of 1985, there has been a continuous tightening of the permissible sulfur content in fuels and of the maximum emissions from power plants. As a consequence, the total annual emission of sulfur dioxide (SO2) has been reduced from 450,000 tons in the seventies to 180,000 tons in 1990. This has had a pronounced effect on the SO2 levels in Danish urban areas. Thus, in Copenhagen, the yearly averages have fallen to about 25%. For nitrogen oxides emitted from the power plants, similar regulations are in force. With this legislation, the most important and crucial source of air pollution in Danish urban areas is road traffic. The contribution of nitrogen oxides from national traffic accounts for nearly half the total Danish emission and is increasing steadily; this is consistent with an observed increase of nitrogen oxides in ambient air. The permissible levels of lead in petrol has been reduced drastically. After an introduction of reduced tax on lead-free petrol, it now accounts for more than two-thirds of the total consumption. As a result, the concentration of lead in urban ambient air has been reduced to less than one-sixth. The introduction of 3-way catalytic converters from October 1990 will result in reductions in the emission of a series of pollutants, e.g., lead, volatile organic compounds, carbon monoxide, and nitrogen oxides. In 1980, a Danish air quality monitoring program was established as a cooperative effort between the authorities, the Government, the counties, the municipalities, and the Greater Copenhagen Council. Subsequently, the program has been revised with greater emphasis on air pollution from traffic. The network now includes monitoring of gasses and particulates, generally at curb side. The effects of planned traffic regulations and measures taken are evaluated by use of air quality models. Only a few studies of the relations between air pollution and health have been carried out in Denmark, but even the fairly modest levels appear to have some impact on sensitive groups, e.g., children and asthmatics. —Environ Health Perspect 102(Suppl 4):55-60 (1994).

Key words: air pollution, air quality, emissions, health effects, monitoring, nitrogenoxides, sulfur dioxide lead

Introduction

The air quality investigations and measures for the improvement of air quality in Denmark are taken care of by the National Environmental Research Institute (NERI) and the National Agency of Environmental Protection (NAEP) in cooperation with the local authorities. In the following, the activities in Denmark within the scope of the conference are described briefly. Greatest emphasis has been attached to the air pollution situation in greater Copenhagen, but general aspects of the air pollution have been illustrated by results from other Danish cities, where data were available.

Emissions

Legislation and Abatement

The Danish air pollution abatement is based mostly on emission control. Since the ratification of the international sulfur protocol of 1985, there has been a continuous tightening of the permissible sulfur content in fuels and of the maximum emissions from

power plants. Thus, the maximum sulfur content in gas oil (typically used for domestic heating and thus of importance in urban areas) was reduced to 0.2% by 1988. For larger power plants (less than 25 MWe), the total emission of sulfur dioxide for 1991 to 1994 has been limited to 620,000 tons by an act of 1989. For nitrogen oxides (calculated as NO₂) emitted from the power plants, a similar quota of 431,000 tons has been fixed.

Emissions of toxic compounds from the industrial sector are regulated according to guidelines (1) negotiated with the Federation of Danish Industries and Danish Employers' Confederation. Limits for mass streams, emissions, and contributions to ambient emissions are indicated for about 200 chemicals. On the basis of these values, it is possible to calculate the stack or release height with an authorized dispersion model.

With this legislation in force, the most important and crucial source of air pollution in Danish urban areas is road traffic. In 1977 the lead content in petrol was 0.55 g/L and the resulting Danish lead emission about 1000 tons/year. Since then, the permissible levels have been reduced drastically. In 1978, they were reduced to 0.40 g/L; in 1982 they were reduced to 0.15 g/L for regular grade; and in 1984, they were reduced to 0.15 g/L for premium grade. Following an introduction of a reduced tax on lead-free (less than 0.013 g/L) petrol, it

now accounts for more than two-thirds of the total consumption.

Since October 1, 1990, emission standards equivalent to the U.S. standard are valid for passenger cars and small vans (payload less than 760 kg). In practice this means that all new gasoline-driven passenger cars are equipped with a closed-loop 3-way catalytic converter. Since the same date, all new heavy-duty trucks (less than 3500 kg) must meet European Economic Community (EEC) directive 88/77. Since July 1, 1991, the use of light diesel (sulfur content below 0.05%) in buses has been promoted by a reduction in tax; since July 1, 1992, this has been extended to all diesel autos.

Past and Present Emissions

As a consequence of the regulations the total emission of sulfur dioxide has been reduced from 450 kt per year in the seventies to 180 in 1990 (Figure 1). The most pronounced reductions in relative terms have taken place for individual domestic heating systems (50–9 kt) and for district heating (61–16 kt). The reasons behind these reductions are more efficient systems, better insulation of buildings, and use of natural gas. Further, combined heat-power generation has come into use. This has had a pronounced effect on the SO₂ levels in Danish urban areas.

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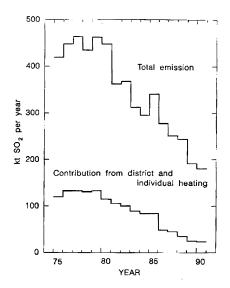


Figure 1. Total national Danish sulfur dioxide emissions (2). The contribution from district and individual domestic heating is also shown separately. A roughly constant contribution from international traffic (24 kt in 1990) is not included.

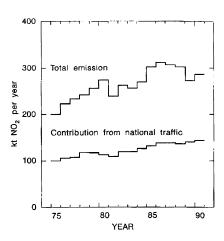


Figure 2. Total national Danish emissions of nitrogen oxides, calculated as nitrogen dioxide (2). The contribution from national traffic (mainly road traffic) is also shown separately. A roughly equal contribution comes from power plants. The remaining part, which has declined from 20 to 10%, comes from domestic heating and industry. In addition, there is a contribution from international traffic (about 78 kt in 1990); this, however, has little bearing on urban air quality.

For nitrogen oxides, on the other hand, there have only been some shifts between contributions from various sectors but no significant overall reduction. Figure 2 shows the development in nitrogen oxides (NO_x) emission since 1975. The variations in the total emission are due to varying heating demand and import of electricity. The contribution from national traffic accounting

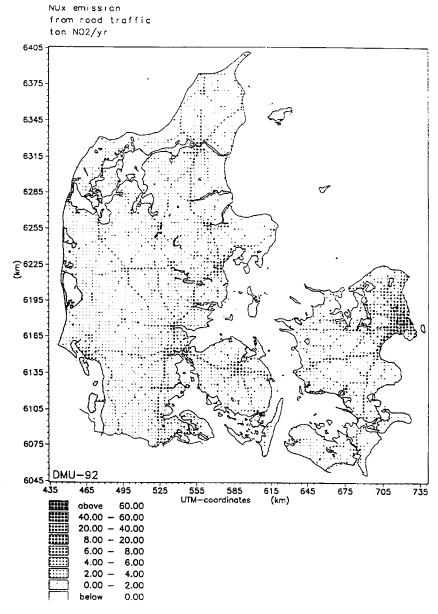


Figure 3. Spatial distribution of nitrogen oxide emissions from road traffic in Denmark in 1985 (3). The actual calculations are performed in a 1 x 1 km grid. Unit ton NO_2 /km per year. Note that the scale is not linear.

for nearly half of the emission, however, is steadily increasing by about 1% per year. As demonstrated in Figure 3, the emission is concentrated in cities and along main roads. Notwithstanding the complex oxidation of nitric oxide to NO₂, this is consistent with the observed increase in NO_x concentrations in Danish urban areas (see later).

Danish emissions of volatile organic compounds (VOCs) have not been studied systematically and no time series exist. In 1985 the total emission of nonmethane VOCs was estimated to about 2000 kt/year with main contributions from domestic road transport (43%) and use of solvents (30%) (2). Some reduction in the industrial

sector must be assumed due to changes in technology, e.g., more extensive use of water-based paints. Only in specific cases, e.g., dioxins from incineration plants, have individual compounds been identified (4).

Planned Development

In 1988 the Danish government published a general action plan (5) to follow up on the recommendations on the report of the World Commission on Environment and Development (6). Subsequently, the Ministry of Energy presented a specific plan concerning energy (7). According to this plan, the following changes from the situation in 1988 will be effected by the

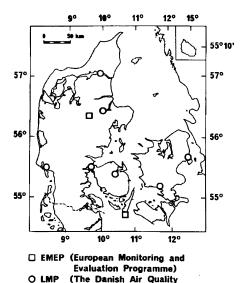
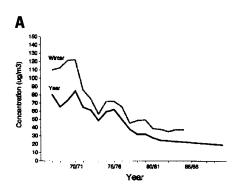


Figure 4. Map of Denmark with location of monitoring stations in EMEP and indication of urban areas included in the Danish Air Quality Monitoring Program.

Monitoring Network)



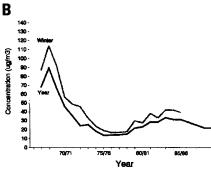


Figure 5. Annual average concentrations of sulfur dioxide (*A*) and black smoke (*B*) in the center of Copenhagen measured in the period 1966 to 1991.

year 2005: a) the gross consumption of energy will be reduced by nearly 15%; b) the consumption of natural gas and renewable energy will be increased by about 170 and 100%, respectively; and c) the consumption of coal and oil will be reduced by

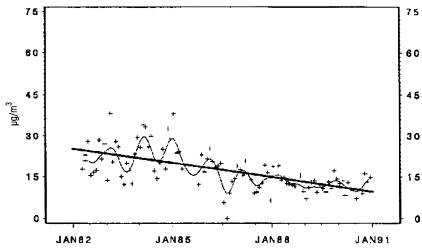


Figure 6. Individual monthly averages, smoothed curve, and linear regression line of sulfur dioxide concentrations measured in Aalborg, 1982 to 1991.

about 45 and 40%, respectively. The consequences for total Danish emissions will be reductions of about 20% for carbon dioxide, 60% for sulfur dioxide, and 50% for nitrogen oxides.

The transport sector was treated in a separate plan (8). Here, the following goals are presented: a) a stabilization of energy consumption and carbon dioxide emission before the year 2005 and a reduction of 25% by the year 2030; b) a reduction in the emissions of nitrogen oxides and volatile hydrocarbons of at least 40% before the year 2000, of 60% before the year 2010, and more up to the year 2030; and c) a halving of the particle emission in cities up to the year 2010 and further reductions up to the year 2030. In none of these plans are specific environmental benefits calculated, but if the intentions are carried out, they will-irrespective of efforts in other countries-have a substantial impact in urban areas, where the pollution is mainly of local

Monitoring Programs

Air quality monitoring started in 1966 by the Greater Copenhagen Air Pollution Board and continued until the late 1970s, partly in cooperation with the Air Pollution Laboratory (under NAEP). In 1980 a Danish Air Quality Monitoring Program was established as a cooperative effort between the authorities of the government, counties, municipalities, and the Greater Copenhagen Council. The program (Figure 4) was carried out in seven Danish cities including greater Copenhagen (9,10). In addition to the monitoring, the program included emission inventories and

dispersion calculations for sulfur dioxide (11,12). In 1987 and 1991 the program was revised with greater emphasis on air pollutants from the traffic. The network now includes monitoring of gasses and particulates, generally at curb side. Data from the monitoring stations are transmitted directly to NERI and to the air pollution unit of the Greater Copenhagen Region.

The effect of planned traffic regulations and abatements can be evaluated by use of reliable air quality models. For that purpose, NERI has developed advanced street pollution models that incorporate both dispersion and chemical reactions in urban areas (13-15). The main objectives of the air quality monitoring program are to: a) determine any times air quality limit values or guidelines were exceeded; b) show trends of the air quality; c) give information about the air quality to the public; d) identify air pollution sources; and e) supply data to the research within the field of air pollution.

The Measured Air Quality in Danish Cities

Air Pollution from Stationary Sources

Extensive time series have only been recorded for sulfur dioxide and black smoke. Data from one station in the center of Copenhagen show that the concentration of sulfur dioxide as well as black smoke increased until 1972 (the first energy crisis) and then decreased until the late seventies (Figure 5A,B). This negative trend is due to several factors: energy saving, more district heating (with tall stacks instead of small chimneys), generally higher stacks, limits on sulfur content in fuel, increasing use of natural

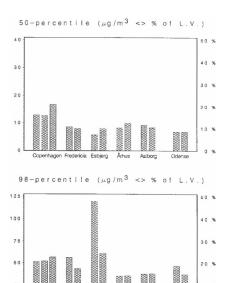


Figure 7. Fifty and 98% of sulfur dioxide concentrations at 13 monitoring stations in Danish urban areas in 1990. At the right axis, the percentage of the Danish limit value is marked.

gas, and more efficient combustion. Today, district heating covers more than 90% of residential and occupational heating in many Danish urban areas.

From the late 1970s until today, the sulfur dioxide concentration in most Danish cities, including greater Copenhagen, has shown a slightly negative trend (Figure 5A,6). On the other hand, the black smoke concentration has been increasing until the late 1980s in the center of Copenhagen (Figure 5B). Receptor model calculations indicate that this is probably due to an increasing contribution from traffic and long-range transport of air pollution (16). In the last few years, the air pollution in Danish cities has generally been low and the seasonal variation weak due to mild winters.

Generally the levels of sulfur dioxide are low compared to the EEC limit values. Only in Copenhagen has the annual average in some years been near the EEC guideline (40 µg/m³) (9,10) (Figure 7).

Air Pollution from Traffic

In the eighties the most important air pollution source in Danish cities was road traffic: vehicles, buses, and lorries. The pollutants are carbon monoxide, nitrogen oxides, particulates, and toxic compounds in the particulates (heavy metals and organics) and in the gaseous phase (organics). Time series for approximately 10 years at the same monitoring stations exist for lead and nitrogen oxides.

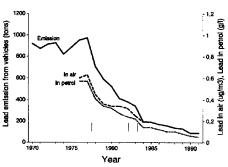


Figure 8. The development of lead concentration in the air in Copenhagen. In the same graph the lead content in petrol and the total lead emission from vehicles are shown. The bars indicate a tightening of the permissible lead content in petrol (1978: 0.40 g/L; 1982: 0.15 g/L standard; 1984: 0.15 g/L premium). After a later introduction of reduced tax on lead-free (less than 0.013 g/L) petrol, it accounts for about two-thirds of the consumption.

Because more than 80% of the lead was emitted to the air from petrol-driven vehicles, a very clear correlation between lead content in petrol, total lead emission in Denmark, and lead concentration in the air was observed (Figure 8). The lead concentration in the air was well below the EEC limit value (yearly average 2 μ g/m³). The lead concentration will decrease even more in the years to come due to introduction of 3-way catalysts in all new vehicles from October 1990.

Nitrogen monoxides have been monitored continuously since 1982 in most of the largest cities in Denmark. In the center of Copenhagen, the EEC guidelines for nitrogen dioxide (135 µg/m³ as 98 percentile of 1-hr average and 50 µg/m³ as 50 percentile of 1-hr average) were generally exceeded. The EEC limit value (200 µg/m² as 98 percentile of 1-hr average) was not exceeded at the monitoring stations (Figure 9). Figure 10 shows the average concentration of nitrogen monoxide and nitrogen dioxide in the center of Aalborg. The measurements have shown a slightly positive trend in Danish cities. The main contribution to nitrogen monoxide in streets is the traffic. The nitrogen monoxide concentration is much lower behind the buildings along the street than at the curb side, where as nitrogen dioxide concentration is much more evenly distributed in the center of the urban areas. This is due to the fact that nitrogen dioxide is formed by oxidation, particularly by ozone, of nitrogen monoxide within less than 1-hr. Only approximately 5% of the nitrogen oxides are emitted as nitrogen dioxide directly from the motor vehicles.

The increases were caused by increasing traffic (a few percent per year since 1980). More than 80% of the nitrogen dioxide in urban air originates from the road traffic. Figure 11 shows an example of diurnal variation of nitrogen monoxide and nitrogen dioxide on weekdays and Sundays; the concentrations are strongly correlated to the traffic density. Consistent with this, the ozone concentration in Copenhagen varies inversely with traffic intensity, being generally high during the weekends and low during rush hours on weekdays (17).

Special investigations of the air pollution in busy streets at the curbside in Copenhagen have indicated that the World Health Organization limit value of carbon monoxide and the EEC limit values of nitrogen dioxide were exceeded in narrow street canyons with 16,000 to 22,000 vehicles per day. Model calculation could be used for identification of such streets. The high and slightly increasing air pollution from the traffic has led to the introduction of 3-way catalysts for all new cars from October 1990. This reduced the emission of nitrogen oxides, carbon monoxide, and organics. The nitrogen dioxide concentration in urban air is not reduced correspondingly because it is formed by oxidation of nitrogen monoxide. Here, ozone put in the region is the limiting factor. As a consequence of the introduction of 3-way catalysts, the vehicles must use lead-free

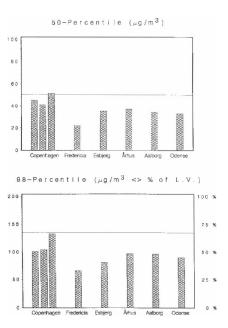


Figure 9. Measured 50 and 98% of nitrogen dioxide at eight monitoring stations in Denmark in 1990. The solid lines are Danish guidelines and the right axis represents percentage of Danish limit values.

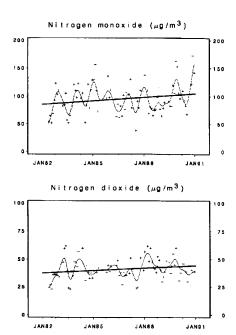


Figure 10. Individual monthly averages, smoothed curve, and linear regression line of the nitrogen monoxide and nitrogen dioxide in Aalborg, 1982 to 1991.

petrol; the lead emission will therefore be further reduced.

Air Pollution Episodes

Air pollution episodes occur occasionally in Danish cities. They may be caused by local buildup of pollution emitted from low sources (e.g., vehicles, domestic heating, and industry) during inversion, but longrange transport also is important. Denmark is located in a region that is strongly influenced by long-range transport of pollutants from the highly industrialized areas of Europe. The air pollution episodes in Denmark therefore often appear in connection with the breakup of smog episodes in eastern and Central Europe, whereby the polluted air is transported to Denmark and southern Scandinavia. Further, the transport of ozone to the area is an important factor in the photochemical oxidation of nitrogen oxides (i.e., formation of nitrogen dioxide).

Air Pollution and Health Effects in Denmark

Only a few studies of the relations between air pollution and health have been carried out in Denmark within the last 15 years. In the period between 1977 and 1980, a statistical analysis was made of the possible relations between short-term air pollution impacts and removal to hospitals due to acute heart and airways diseases in greater Copenhagen (18). The air pollution expo-

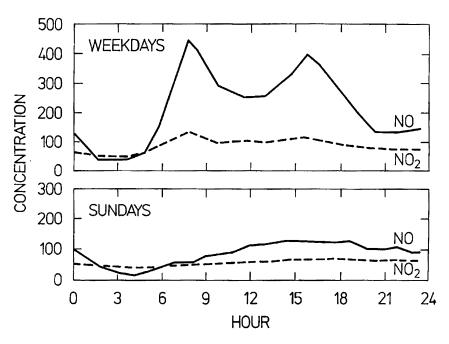


Figure 11. Diurnal variation of nitrogen monoxide and nitrogen dioxide in Copenhagen on weekdays and Sundays. The average concentrations (μ9/m³) of every hour are calculated by measured 1-hr average concentrations within 1 year (1982).

sures were estimated from measurements from fixed monitoring stations in Copenhagen. Although some correlation between the above diseases and the SO₂ and soot pollution was indicated, direct conclusions on the relation between diseases and air pollution exposure were not possible.

The lead content in milk teeth from 1,302 children has been investigated in Århus in Denmark (19) because it is assumed to be an indicator for the integrated exposure from birth. The lead content was tentatively related to the exposure from traffic based on location of day-care institutions and permanent addresses, and it was concluded that children living near busy streets generally had a higher lead content in the milk teeth than children from low exposure areas. Also, the fathers' work place exposure and the mothers' smoking habits during pregnancy influenced the lead content. The mental development of the children was investigated, and some learning disabilities were observed in the group with high lead exposure (19).

From 1988 to 1989, an investigation (20) based on questionnaires compared nuisance and symptoms of workers in busy streets in Copenhagen to those of gardeners working in cemeteries, parks, etc. In the first group, more frequent complaints were observed of headache, dizziness, special taste in the mouth, tendency to fall asleep, and irritations of mucous membranes.

From 1977 to 1988, a diary investigation was carried out on 73 humans with asthma and 71 humans with chronic bronchitis (21). Data from the diaries and peak flow measurements were analyzed by the so-called neural networks method and related to the weather and the air pollution. The conclusions were that SO₂ and NO₂ may have a synergistic effect on the impact of air pollution on humans with asthma or bronchitis.

Recently, an investigation in Copenhagen on children's morbidity related to air pollution has been published (22). The investigation includes literature studies, studies of relations between exposure and diary notes, and studies of correlation between the number of contacts to the emergency medical service and air pollution. The exposure studies showed that although exposure measured by personal dosimeters for NO2 was generally lower (by a factor of 3-4) than the levels measured at the fixed monitoring stations, the measurements at the fixed stations could be used as a measure for the variation in exposure. The main conclusion is that even the relatively low air pollution in Copenhagen contributes a few percent to the morbidity in the respiratory tracts in children.

Conclusions

The long-time series have been shown to be very useful for the evaluation of the air pollution problems and of the effectiveness of abatement policies. It should be emphasized that long-time series at the same locations and of the same methods are very important. In addition, it is our experience that the most efficient way of investigating the air quality is to combine the monitoring with different types of model calculation based on meteorological data and

emission inventories. This combination is also a powerful tool in the evaluation of the measures taken and for future planning.

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